QUARTERLY PROGRESS REPORT

A STUDY OF TUNGSTEN-TECHNETIUM ALLOYS

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by

The Staff of Metallurgy Development Operation
Hanford Laboratories
General Electric Company
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Introduction

Technetium is a sister element to rhenium and has many properties that are similar to rhenium. It is predicted that technetium will have about the same effects on tungsten as rhenium in regard to increase in workability, lowered ductile to brittle transition temperature, and improved ductility.

The objectives of the current work are to recover technetium from fission product wastes at Hanford Atomic Products Operation and reduce to purified metal; prepare W-Tc alloys containing up to 50 a/o Tc; fabricate the alloy ingots to sheet stock, assessing the effect of technetium on workability; and perform metallurgical and mechanical property evaluation of the fabricated alloys.

The initial report in this series described the separation and purification of eight hundred grams of technetium metal powder.

Current Progress

During the past quarter two compacts of 100 per cent Tc powder were prepared for arc melting using isostatic compression at 80,000 psi. The powder and compacts are shown in Figure 1. Arc melting was performed on a water cooled copper hearth with a tungsten electrode in an evacuated chamber backfilled with argon and gettered by first melting zirconium. Total loss in the two nominal 40 gram buttons after five melting cycles was 0.634 grams or approximately 3/4 per cent. Radiation from each button was 4800 mrad including 8 mrad gamma at $1\frac{1}{2}$ inches. Analytical data obtained on the as-reduced powder and arc-melted material are given in Table I. The total impurity content is about 150 ppm.

TABLE I.

Analysis of As-Reduced Technetium Powder and Arc-Melted Metal

Element	Powder 1	Powder 2	Powder 3	Metal 1
	---Values in ppm---			
Oxygen *	1620	1260	413	< 1.7
Nitrogen	60	20	12	3
Hydrogen	44	39	18	< 0.006
Aluminum **	50			
Boron	1-2			
Calcium	5			
Iron	50			
Sodium	20			
Silicon	20			
Strontium	Trace			
Others	Not Detected			

* Vacuum fusion-platinum bath

** Semi-quantitative spectrographic

One of the arc-melted buttons was prepared for metallographic examination and micro hardness measurements. The macrostructure of the as-cast arc melted material shown in Figure 2 is fine grained and not columnar in nature like tungsten. Small inclusions were observed in the polished condition as shown in Figure 3. These are not located at grain boundaries and are not identified. They are presumed to be TcO_2 or Al and Si oxides. The microstructure shown in Figure 4 is typical of the metal in relatively strain free areas and that in Figure 5 of metal with some cold deformation resulting from squeezing the button. A microhardness traverse across the length dimension of the surface shown in Figure 2 indicated hardness variation from DPH 113 to 189. Lower values were generally in strain free areas.

The button was encapsulated in a heavy-walled stainless steel compact by electron beam welding and hammer forged from 1000 C preheat to a height reduction of 42%. X-radiography of the compact revealed no edge cracking. Additional hot rolling was performed from 1000 C preheat to a height reduction of 33%. X-radiography indicated edge cracking at this point and fabrication was stopped for desheathing.

The technetium surface after removal of the sheath was extremely rough with depressions apparently related to the cast grain size. No edge cracking due to fabrication was evident. Part of the surface roughness is probably due to the relative softness of the stainless steel at the rolling temperature. The technetium was reduced to 0.140-inch thickness for a total fabrication reduction of approximately 65 per cent. It is planned to surface condition the material and resheath with thin tungsten plates on each side prior to further rolling.

Two additional 100 per cent technetium melts were made by electron beam melting in a 10 KW EB evaporator unit. Loose powder was used as the charge. The resulting melts were 8.2 and 6.7 grams with losses of 0.2 and 0.4 grams. Metallographic examination is in progress on one of these buttons and an attempt will be made to form the remaining button to a wire.

The processing of the pure technetium metal has been used as a means of establishing the working conditions required for radiation and contamination control. Procedures are now established and a hood has been set up for some operations with the powder. The compaction and arc melting of a series of W-Tc alloys will proceed during the next quarter.

Literature Review

Reference articles concerning the alloying behavior, properties, or fabrication of technetium are listed below and that material of particular interest to the W-Tc study is reviewed.

1. J. B. Darby, Jr., D. S. Lam, L. J. Norton, and J. W. Downey, Intermediate Phases in Binary Systems of Technetium-99 with Several Transition Elements, Journal of the Less-Common Metals, 4(1962)558-563.

2. J. B. Darby, Jr., L. J. Norton, and J. W. Norton, A Survey of the Binary Systems of Technetium with Group VIII Transition Elements, Journal of the Less-Common Metals, 5(1963)397-402.

3. J. B. Darby, Jr., L. J. Norton, J. W. Downey, Technetium Compounds with the $MgZn_2$ Structure, Journal of the Less-Common Metals, 6(1964)165-167.

4. M. G. Chasanov, I. Johnson, and R. V. Schablaske, The System Zinc-Technetium-99, Journal of the Less-Common Metals, 7(1964)127-132.

5. J. Niemiec, X-Ray Analysis of Technetium-Molybdenum Alloys, Bulletin De L'Academie Polonaise Des Sciences, Vol XI, No 6, 1963.

6. J. Niemiec, X-Ray Analysis of Technetium Binary Alloys With Tungsten and Rhenium, Bulletin De L'Academie Polonaise Des Sciences, Vol XI, No 6, 1963.

7. Brazing Technology - W-TcAlloy System, GEMP-35A, May 28, 1964, 32-33.

8. Binary and Ternary Phase Diagrams of Columbium, Molybdenum, Tantalum, and Tungsten, DMIC Report 152, April 28, 1961. (Supplement DMIC Report 183, February 7, 1963)

Figures 6 and 7 show the available data concerning the binary phase diagrams of tungsten with rhenium and technetium. The limited data shown in Figure 7 are from J. Niemiec[(6)] and are based on twelve sintered alloys of 30-50 μgm covering the whole composition range. The major features of the system were reported to be:

1) W α-primary solution; 55 a/o Tc at 1800 C and 33 a/o Tc at 1200 C.

2) δ phase; 70 and 75 a/o Tc alloys were single phase δ when quenched from 1200 C.

3) TcΘ-final solid solution; 12 a/o W at 1800 C and 7 a/o W at 1200 C.

No study was made of the liquid-solid reactions. The major difference between the preliminary W-Tc diagram and that established for W-Re[8] is that only one intermediate phase was identified in the W-Tc system where two intermediate phases exist in the W-Re alloy system. It is interesting to note that Tc has higher solubility in tungsten than does rhenium in the range 1000-1800 C.

Recent work at NMPO[7] with W-Tc alloys ranging from 9 to 19 w/o Tc (16 to 30 a/o) showed that these sintered alloys clad in molybdenum were cracked during hot rolling at 1350 C to a total reduction in thickness of 50 per cent. Arc melted buttons of the same composition were brittle during cold forging to 50 per cent reduction in thickness.

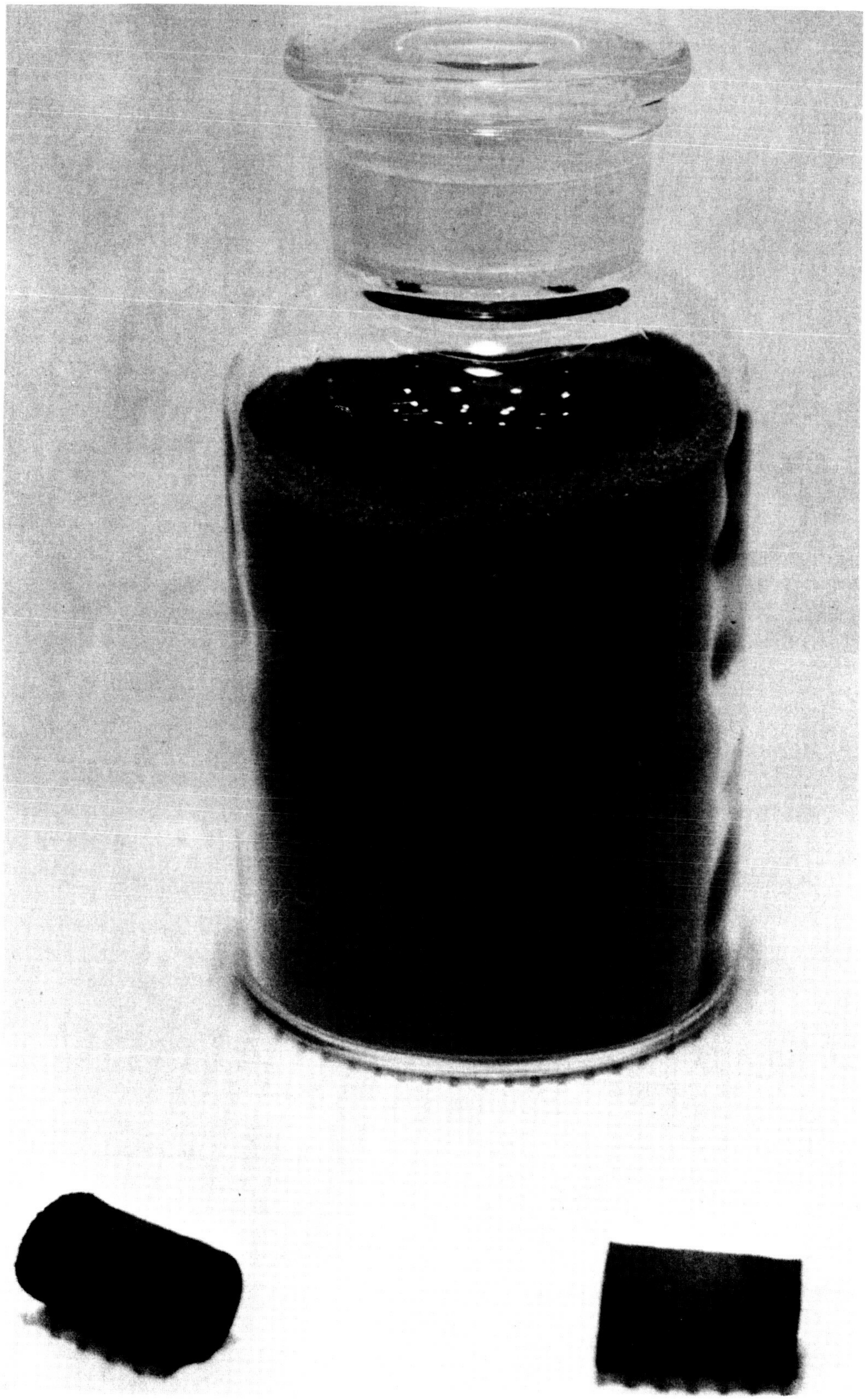


FIGURE 1

800 Grams As-Reduced Technetium Powder. Two 40 Gram Isostatically Compressed Compacts.

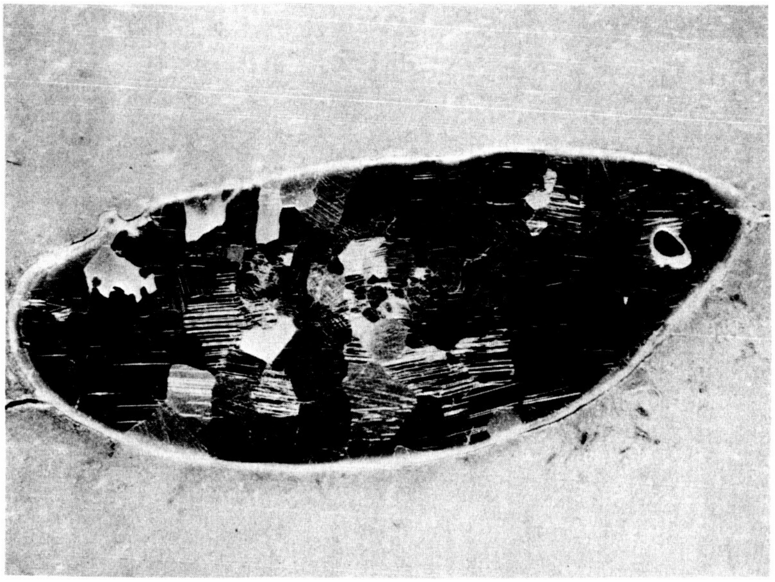


FIGURE 2

Macrostructure of Arc Melted Technetium Button - 10X

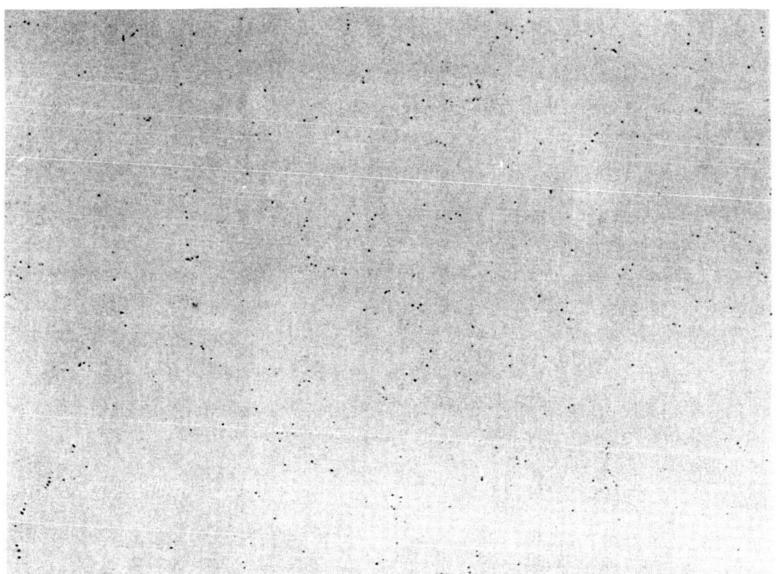
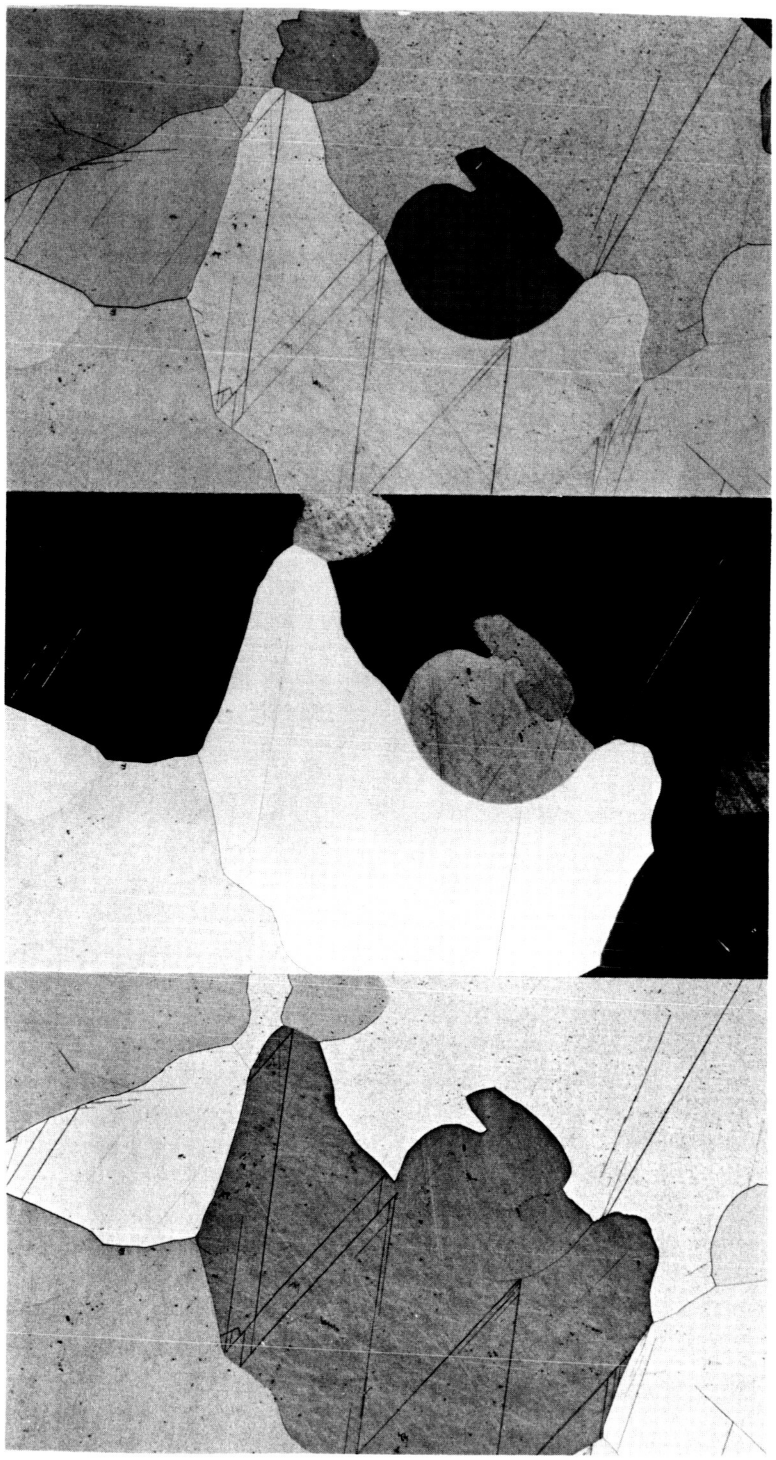


FIGURE 3

Arc-Melted Technetium -As Polished
100X. Showing Inclusions.



Sensitive Tint

Polarized Light

Bright Field

FIGURE 4

Microstructure of Arc-Melted Technetium.
All 100X. 10% Oxalic Acid Etch
3 Seconds at 6 Volts.

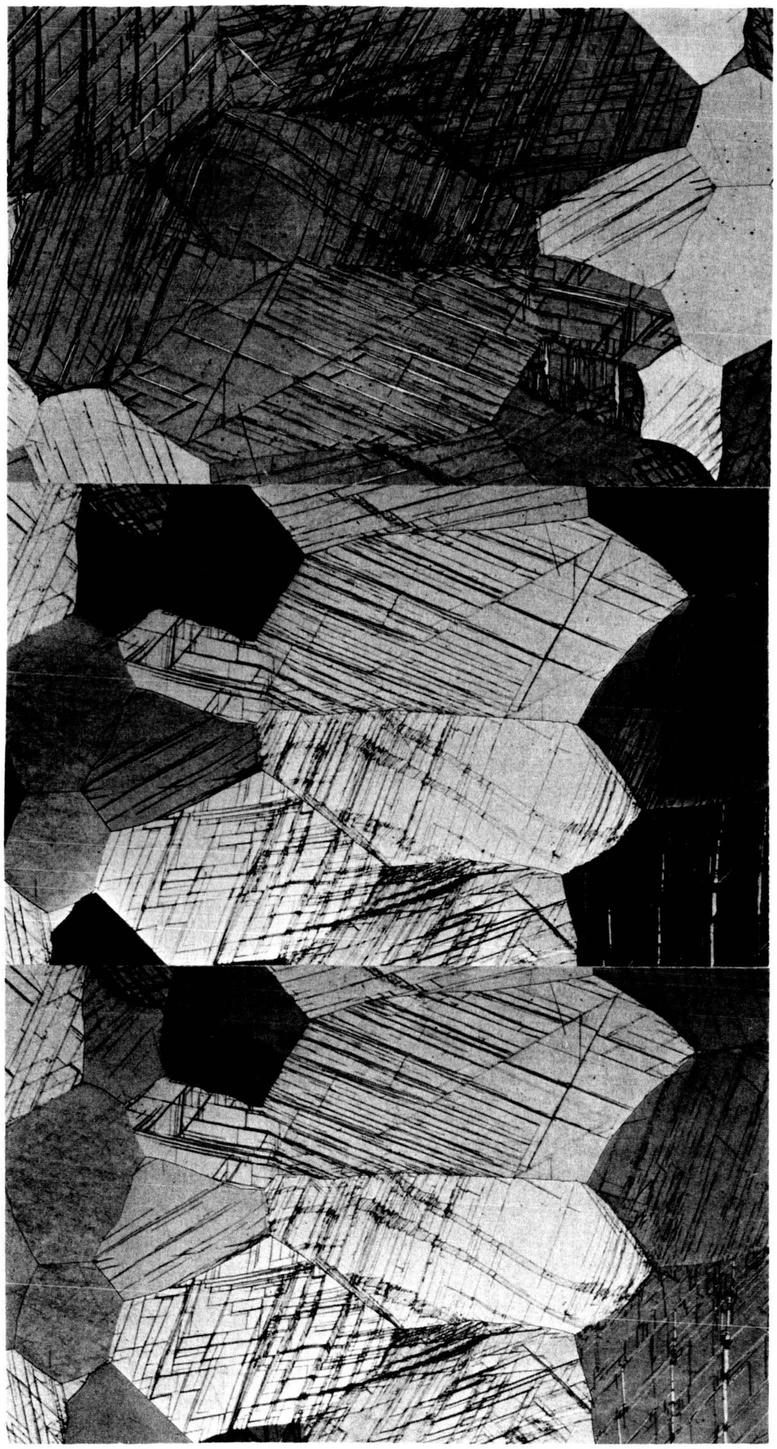


FIGURE 5
Microstructure of Arc-Melted Technetium
All 100X. 10% Oxalic Acid Etch
3 Seconds at 6 Volts.

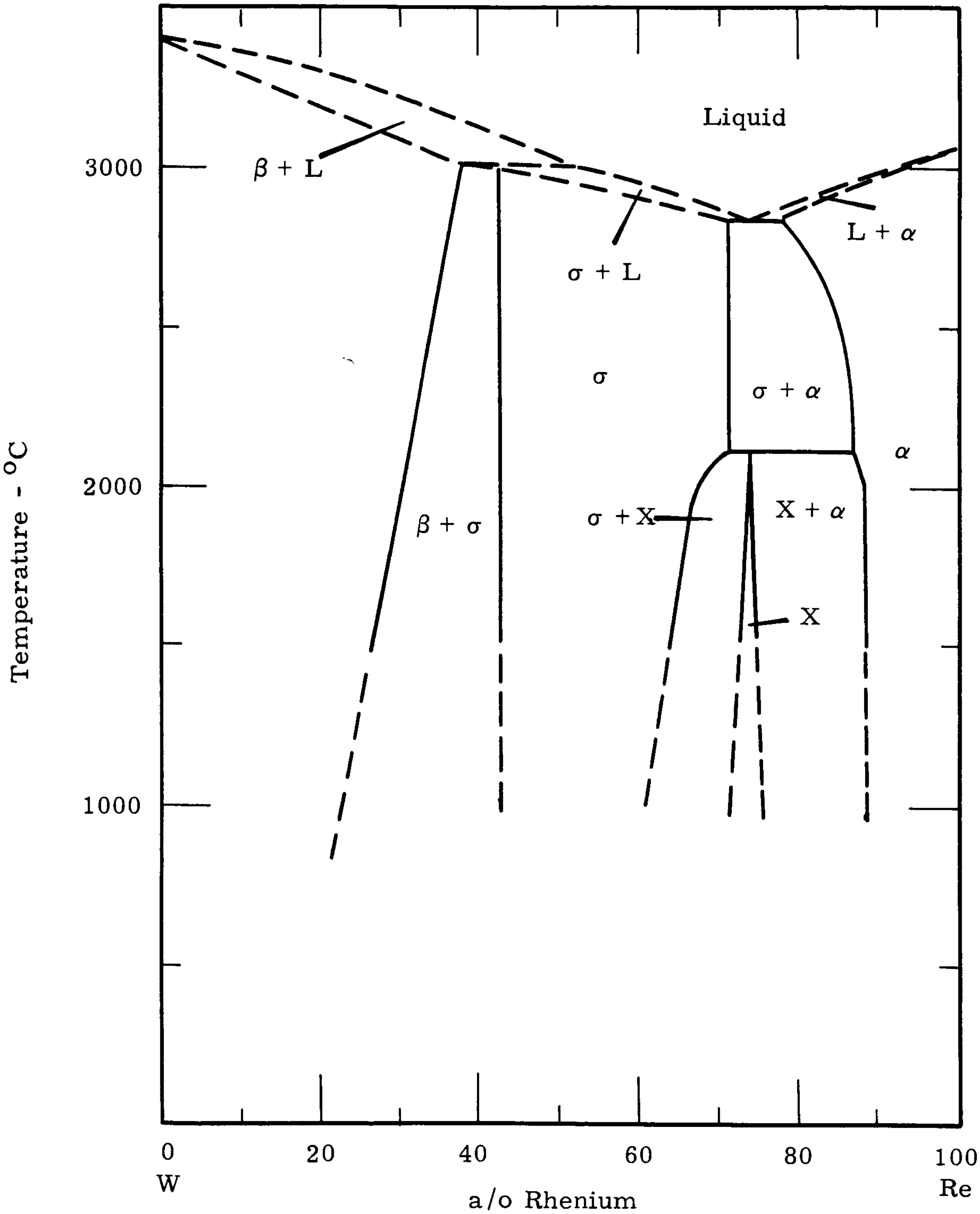


FIGURE 6

Tungsten-Rhenium Binary Phase Diagram.(8)
(DMIC 152)

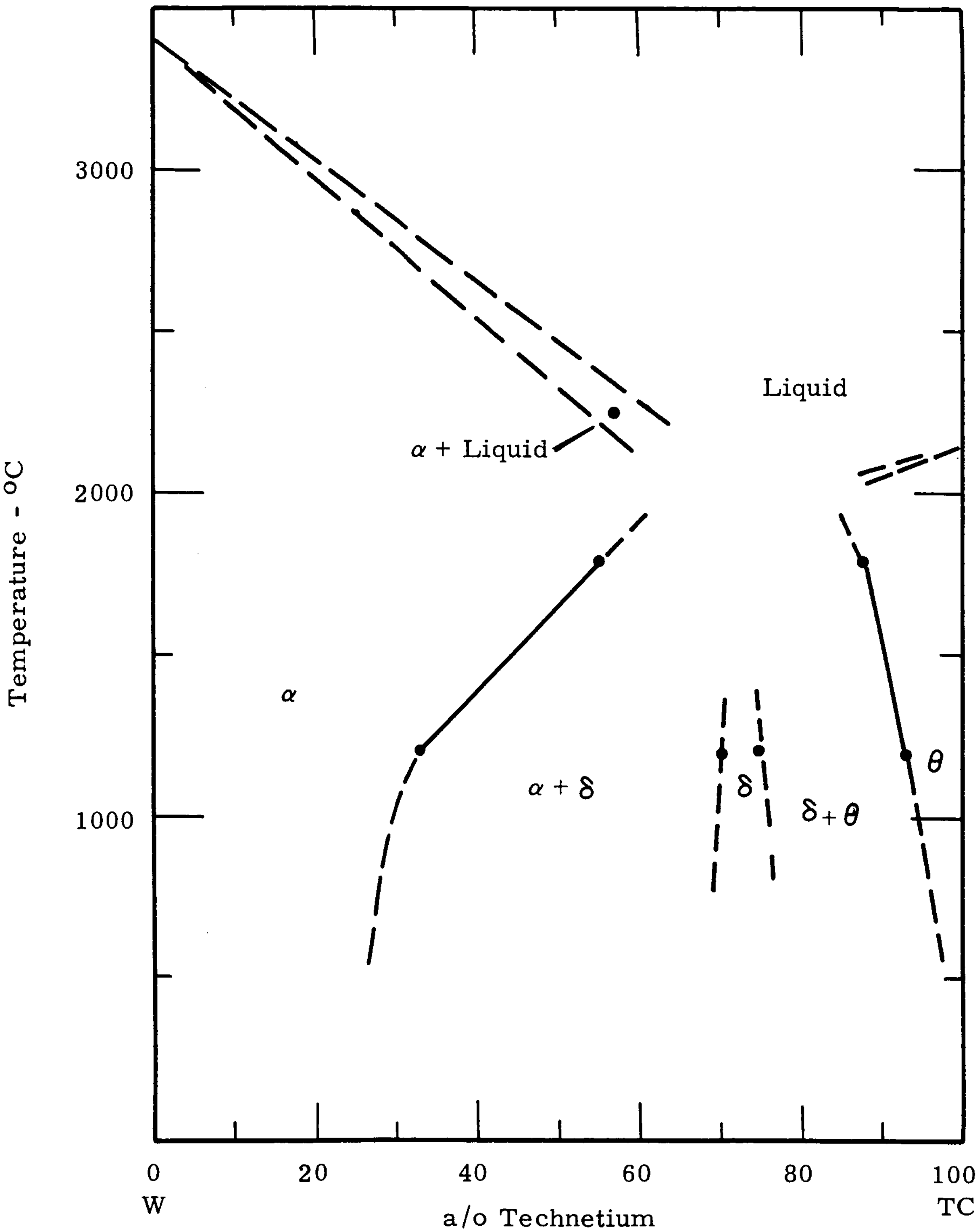


FIGURE 7

Tungsten-Technetium Binary Phase Diagram.(6)